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(54) **THERMAL CYCLE RECIRCULATING PUMP FOR ISOTOPE PURIFIER.**

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**US-A- 2 533 966**  
**US-A- 3 144 200**  
**US-A- 3 758 777**

**NUCLEAR INSTRUMENTS AND METHODS,**  
vol. 139, 1976, pages 101-104, North-Holland  
Publishing, Co., Amsterdam, NL; R. MEUNIER  
et al.: "Progress report on separators Sidon-  
ie and Paris"

**PATENT ABSTRACTS OF JAPAN, vol. 8, no.**  
**258 (M-340)[1695], 27th November 1984; &**  
**JP-A-59 131 070**

**R. Meunier et., "Progress Report on Separa-  
tors Sidonie and Paris", Nuclear Instruments  
and Methods, 139 (1976), see pages 101-104,  
North Holland Publishing Co.**

(73) Proprietor: **HILL, David A.**  
**18522 Wells Drive**  
**Tarzana, California 91356(US)**

(72) Inventor: **HILL, David A.**  
**18522 Wells Drive**  
**Tarzana, California 91356(US)**

(74) Representative: **MEISSNER, BOLTE & PART-  
NER**  
**Patentanwälte**  
**Postfach 86 06 24**  
**D-81633 München (DE)**

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## Description

The invention relates to a system for the separation and purification of a selected isotope from a polyisotopic mixture.

This invention will separate and enrich a single isotope such as iodine-123, to very high purity, from a polyisotopic mixture produced by a conventional small cyclotron. This invention will also greatly increase the recovery rate of the selected isotope, thus lowering the cost of pure isotopes, and making feasible their use in nuclear medicine.

The invention accordingly comprises the several steps and the relation of one or more of such steps with respect to each of the others, and the apparatus embodying features of construction, combinations of elements and arrangements of parts which are adapted to effect such steps, all as exemplified in the following detailed disclosure, and the scope of the invention will be indicated in the claims.

## BACKGROUND OF THE INVENTION

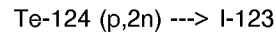
Many elements have chemically identical isotopes, which vary only in the number of their neutrons. These isotopes of a single element are often co-produced in a single reaction. The isotopes of a single element do vary, particularly in their radioactivity, and therefore purification or separation of the isotopes leads to an improved product. However, these isotopes, being chemically identical, are difficult to separate.

By way of example, iodine-123 is a close-to-ideal radioactive isotope used in nuclear medicine. It permits diagnostic tomography of a patient's brain to allow a physician to distinguish between multiple infarct dementia, associated with a series of strokes, and Alzheimer's Disease. These two diseases, requiring quite different treatment, have, prior to diagnostic tomography, given only obscure and conflicting symptoms.

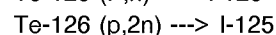
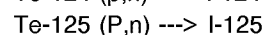
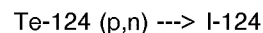
Pure iodine-123 is close-to-ideal because it has a 13 hour half life allowing shipment nationwide and yet decaying sufficiently rapidly so that the patient does not receive an excessive dosage of radiation. Pure iodine-123 emits a single characteristic energy, 159 keV, and practically no other radiation. Pure iodine-123 is well tolerated by the human organism and is readily compounded as a label into many biochemical agents without disrupting their action.

The principal drawback to the use of pure iodine-123 in nuclear medicine is that pure iodine-123 is expensive to produce. Most of the iodine-123 which is available is not pure. It is produced by a 20 to 40 MeV cyclotron or linear accelerator. The high energy protons from the cyclotron bombard a

target enriched in tellurium-124, whose atomic number is 52 compared with iodine, whose atomic number is 53. The high energy proton principally dislodges two neutrons to produce iodine-123 in the reaction:

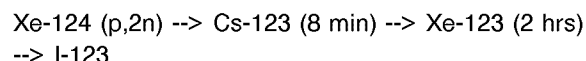


Some of the bombarding protons dislodge only one neutron. In addition, the target contains residual amounts of tellurium-125 and tellurium-126. The result is that about 3% of iodine-124 and iodine-125 is co-produced in the side reactions:



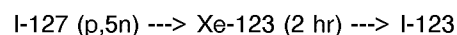
These two isotopes of iodine, even a few percent, are each unwanted in nuclear medicine for different reasons. Iodine-124 emits other radiation which degrades the output of diagnostic tomography, making it a less precise test. The radiation from iodine-125 is soft, i.e., X-rays which are absorbed within the patient, and therefore does not degrade the output of diagnostic tomography. However, iodine-125 has a half life of 60 days, thus substantially increasing the internal radiation dosage to the patient.

Two alternative processes exist to produce pure iodine-123. The first one uses a 20-40 MeV cyclotron in which the high energy proton bombards a target of xenon-124 and dislodges two neutrons to produce cesium-123. This then decays in 8 minutes to xenon-123, which decays in 2 hours to iodine-123, according to the reaction:

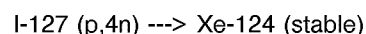


This reaction produces high purity iodine-123, but does so very expensively because xenon-124 is a very rare isotope. The process was, until recently, therefore limited to research applications.

The second process uses a 70 MeV cyclotron with an iodine 127 target the high energy proton can dislodge 5 neutrons to produce xenon-123, which decays in 2 hours to a solid iodine-123 according to the reaction:

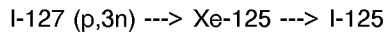


In a first side reaction the proton dislodges 4 neutrons to produce a gaseous xenon-124, according to the reaction:



which is a stable gas, and further separates from the solid iodine-123.

Another side reaction occurs. In this, the proton dislodges three neutrons to produce xenon-125, which decays to iodine-125, according to the reaction:



About 0.2% of iodine-125 is co-produced and is a contaminant.

To date, all 70 MeV cyclotrons have been built for research, not commercial applications, due to the expense of building and operating them. One 70 MeV cyclotron became operational in the United States in 1987, and will be the first one dedicated to the commercial production of radioactive isotopes for nuclear medicine.

A technology to purify or separate a polyisotopic mixture has existed since the 1920's. This is the magnetic mass spectrometer or mass analyzer, which achieves a physical separation according to mass by differentially deflecting ionized isotopes of different atomic weight, the lighter isotopes being deflected more than the heavier isotopes.

The first step in a magnetic mass spectrometer is to ionize the isotopes in an ion source. At most only a small portion of the isotopes can be ionized and the remainder of the isotopes escape into the vacuum chamber with no charge and are therefore neither accelerated by the electrode nor deflected by the magnet. Recoveries of 1% to 15% of the desired isotope are typical.

This low recovery rate is acceptable in an analytical or research magnetic mass spectrometer. This technology becomes uneconomic, however, for commercial applications, including separating radioactive isotopes for nuclear medicine.

These elements of the present invention appear in innumerable prior art publications, and applicant will not attempt to separately identify what may be the closest prior art.

The applicant has found only two items in the prior art pertinent to the present invention:

Simmons, U.S. Patent No. 2,533,966, issued December 12, 1950, discloses a novel method to accelerate ions, a moving magnetic field. Simmons also discloses an ion source 17, having two feed pipes for ions, 13 and 14, and two exit pipes 19 and 22. Pipe or casing 22 contains a focusing field 45 for the ionized isotopes to exit into the magnetic field of force 48 of a mass spectrometer. At the other end of the ion source is a grid 20 which repels the ionized isotopes but allows the un-ionized isotopes to enter pipe or conduit 19. These isotopes, supplemented by the feed stock, are recirculated to the ion source through pipes 13 and

14. There is no means to prevent un-ionized isotopes from escaping at the larger opening 21 nor any recirculation of any un-ionized isotopes which escape the ion source 17 at opening 21.

There are crucial differences between the Simmons disclosure and the present invention. First, the present invention recirculates the uncharged isotopes which have left the ionizer chamber with the charged ions, while Simmons withdraws uncharged isotopes from the ionizer and merely recirculates them to the ionizer with no apparent means for increasing the recovery rate thereby. Secondly, the present invention recirculates by first adsorbing and then desorbing the iodine isotope on a glass surface. Simmons recirculates the isotope in the same gaseous state in which it left the ionizer.

Meunier, et al., Nuclear Instruments and Methods 139 (1976) 101-104 discloses a "closed loop circuit" used on two research machines for isotope separation near Paris, France. The machine has a conventional ion source, aperture, vacuum chamber and magnetic mass spectrometer.

Spaced along the vacuum chamber are a series of side chambers, each containing a vacuum pump for the un-ionized isotopes. There is a cascade series of pumps, including oil diffusion pumps, which raise the pressure from  $10^{-5}$  Torr to atmospheric pressure. The recovered isotopes are mixed with feed stock and reintroduced to the ion source. The overall recovery rate ranges from 11 to 32%, perhaps a four-fold improvement over conventional, non-recovery isotope separators. This system is limited to inert gases which will not bond to the metal pump parts.

The present invention is an internal recycle under vacuum, while Meunier is an external recycle utilizing several cascaded pumps to raise the isotope to atmospheric pressure, then reintroduce it to the ionizer under vacuum. The present invention has a recycle loop which is clean, non-reactive, and therefore suitable for iodine. It is non-contaminating, simple, compact and relatively cheap with a recovery rate which may approach 100%.

These advantages are obtained by a separation and purification system for selected isotopes from a polyisotopic mixture and a method therefor in accordance with claims 1 and 6, respectively. Meunier's recycle loop introduces impurities, is suitable only for inert gases, is complicated, bulky, and expensive. The best utilization reported is 32%.

#### BRIEF DESCRIPTION OF THE DRAWINGS

The invention will be better understood and the objects other than those set forth above will become apparent when consideration is given to the following detailed description thereof. Such de-

scription makes reference to the annexed drawings wherein:

Figure 1 is a schematic view of the elements of the invention;

Figure 2 is a cross-sectional view of first embodiment of this invention;

Figure 3 is a cross-sectional view of a second embodiment of this invention during the first phase of its operating cycle;

Figure 4 is the same cross-sectional view as Figure 3 during the second phase;

Figure 5 is a cross-sectional view of a third embodiment of this invention.

Figure 6 is a cross-section of a fourth embodiment of this invention.

## DESCRIPTION OF THE PREFERRED EMBODIMENTS

### Overview

This invention is disclosed broadly in Figure 1, which shows the elements in a schematic or diagrammatic drawing. Not shown is the prior art cyclotron, for instance 20-40 MeV, which may produce the feed material as a polyisotopic mixture.

The feed material is introduced, through an air lock 90, to the discharge region 59 of the ion source 50. The pressure here is about 0.133 Pa ( $10^{-3}$  Torr). Here ionized isotopes are generated by means to bombard the polyisotopic mixture, such as an electrodeless RF discharge 60. Both ionized isotopes and uncharged isotopes leave the discharge region 59 through an aperture 58 of about 1 mm. diameter in the wall 95 of the discharge region 59.

The singly charged and doubly charged isotopes are accelerated as they leave the aperture 58 under the influence of extraction electrode 92. They form a collimated ion beam 61 which passes through a second aperture 99 in a second wall 88. This portion of the apparatus is maintained at  $1.33 \times 10^{-3}$  Pa ( $10^{-5}$  Torr) by a vacuum pump 91.

The apparatus described above is essentially a prior art ion source 50 for an analyzer such as a magnetic mass analyzer 54, or an electrostatic deflection analyzer or an RF quadrupole analyzer. It will ionize only about 5% to 20% of the isotopes, so the great majority of the particles leaving the prior art ion source 50 are uncharged isotopes.

In the conventional magnetic mass analyzer 54, the uncharged atoms will strike the walls 89 because they are not accelerated and collimated by the extraction electrode 92. The walls of the vacuum chamber in a conventional analyzer are at a temperature which will adsorb, condense or react with and bind any atom of the feed material which strikes them. This invention includes maintaining

the walls 89 of the vacuum chamber 52 at an elevated temperature, namely above the temperature at which the gaseous isotope will adsorb or condense. This temperature is 600 °C.

This invention also includes a thermal cycle recirculating pump 62 which is connected to the vacuum chamber 52 through an intake valve 142. The thermal cycle recirculating pump 62 is also connected through an exhaust valve 143 to the discharge region 59. One or more surfaces of the thermal cycle recirculating pump 62 are alternately heated and then cooled.

During the first phase the intake valve 142 is open and the exhaust valve 143 is closed. One surface of the thermal cycle recirculating pump 62 is cooled to 200 °C. The uncharged isotopes will bounce off the walls 89 of the vacuum chamber 52. At some point they will randomly enter the thermal cycle recirculating pump 62 where they will adsorb or condense on the cooled surface.

Adsorption and condensation both refer to the devaporization of a gaseous substance. Adsorption refers to the substance, such as iodine, combining with another substance, such as the glass wall. Condensation means the liquification of iodine by iodine. Depending on the conditions, either or both processes may occur. These processes are therefore collectively called devaporizing and attaching of the gaseous iodine to the wall of the pump.

During the second phase, the intake valve 142 is closed and the exhaust valve 143 is opened. The thermal cycle recirculating pump 62 is raised to an elevated temperature sufficient to desorb or vaporize the uncharged isotopes and drive them back again into the discharge region 59.

When the discharge region 59 achieves a 20% ionization rate then each atom will, on the average, recirculate from the discharge region 59 through the thermal cycle recirculating pump 62 four times before exiting the ion source 50 as an ionized isotope. This means that the discharge region 59 will draw only 20% new feed material from the cyclotron and the remainder 80% of recycled ionized isotopes from the thermal cycle recirculating pump 62.

The remainder of the elements shown in Figure 1 are a conventional prior art magnetic mass analyzer 54. This includes a conventional, 90 ° deflection, double focusing magnet 63 which bends the collimated ion beam 61 differentially based on the isotopic weight and the charge. The few doubly charged isotopes will strike the inner wall of the vacuum chamber 52 in the vicinity of the magnet 63 and be recovered by the thermal cycle recirculating pump 62.

The singly charged isotopes will be directed to two or more collectors 64 where they condense. One of the collectors 64 is reserved for the ionized

isotope iodine-123. The collectors 64 are withdrawn through a vacuum lock 56.

This apparatus will thus separate the radioactive isotope iodine-123 at a very high purity and at a very high recovery rate.

#### Single Chamber Pump

A first embodiment of the invention is disclosed in Figure 2, which shows the thermal cycle recirculating pump 62 portion shown in Figure 1.

The thermal cycle recirculating pump 62 is mounted vertically above the vacuum chamber 52. It comprises an intake pipe 141, a cylindrical chamber 140, and a return pipe 114. Between the intake pipe 141 and the cylindrical chamber 140 is an intake valve 142. Between the cylindrical chamber 140 and the return pipe 114 is an exhaust valve 143.

Each valve has a valve disc 144 containing an iron armature 121. The valve discs 144 are selectively raised by the first magnet 145 and second magnet 146. When the magnet is removed or de-energized, the valve discs 144 fall by gravity onto the two circular shoulders 147, which each form a valve seat 148.

Both the first magnet 145 and the second magnet 146 have a pair of pole tips 94 that are elongated in the vertical direction. When either first magnet 145 or second magnet 146 is energized, the corresponding valve disc 144 rises off the valve seat 148 and rotates into the vertical plane, thus opening the valve.

The thermal cycle recirculating pump 62 has a heat transfer coil 150 through which gases are cyclically pumped at 600°C and 200°C thus heating or cooling the walls 95 of the heat transfer coil 150.

In the first phase, the first magnet 145 is energized opening the intake valve 142. Cooling gas is pumped through the heat transfer coil 150. The randomly moving uncharged isotopes in the vacuum chamber 52 enter the thermal cycle recirculating pump 62 and are adsorbed or condensed on the surface of the heat transfer coil 150.

In the second phase, the first magnet 145 is de-energized, closing the intake valve 142. The second magnet 146 is energized opening the exhaust valve 143. Hot gases are pumped through the heat transfer coil 150. This desorbs or vaporizes the uncharged isotopes and sends them through the return pipe 114 to the discharge region 59.

#### Dual Chamber Pumping

Figures 3 and 4 disclose a second embodiment of the thermal cycle recirculating pump 62

portion shown in Figure 1. Figure 3 shows a portion of the vacuum chamber 52 between the discharge region 59 and the magnet 63 of the magnetic mass analyzer 54. Along the axis of the vacuum chamber 52 is the collimated ion beam 61 which has been accelerated by the extraction electrode 92. This collimated ion beam 61 proceeds through the second aperture 99 to the magnet 63 of the magnetic mass analyzer 54 as shown in Figure 1.

The outer wall 89 of the vacuum chamber 52 forms a symmetrical cylinder around the central axis of the vacuum chamber 52. The outer wall, as shown in section in Figure 3, has a central section 111 of reduced diameter and two outer sections 112 of increased diameter.

Within the vacuum chamber 52 is cylindrical sleeve 110 which is in sliding and sealing engagement with the central section 111 of the outer wall 89. Embedded within the cylindrical sleeve 110 is an iron armature 121. The cylindrical sleeve 110 forms the inner wall of, alternatively, two annular chambers 113, the outer walls of which are the outer sections 112 of the outer wall 89. The walls of the vacuum chamber 52, the cylindrical sleeve 110 and the return pipe 114 are all made of high purity fused quartz.

Extending radially from each outer section 112 is a return pipe 114 containing an exhaust valve 115. Each exhaust valve 115 is selectively operated as described below. The two return pipes 114 join beyond the two exhaust valves 115 to form a single return pipe 114 connected to the discharge region 59.

On the left is shown a cylindrical square valve seat 116 which both forms a wall of the annular chamber 113 and cooperates with the square end of the cylindrical sleeve 110 to seal off the first pumping chamber 118 forming an intake valve. On the right is the outer section 112 of the outer wall which is beveled. Inside it is a small cylindrical beveled valve seat 117 which cooperates with the beveled end of the cylindrical sleeve 110 to seal off the second pumping chamber.

Outside and surrounding the central section 111 are a first cylindrical magnet 119 and a second cylindrical magnet 120. Inside the cylindrical sleeve 110 and completely encased in high purity fused quartz is an iron armature 121, which cooperates with the cylindrical magnets 119, 120. These are spaced apart the distance of the travel of the cylindrical sleeve 110.

Surrounding the two annular chambers 113 are two series of axially aligned heat transfer fins 122. The heating means 123 directs air at 600°C and, alternately, the cooling means 124 directs air at 200°C onto the fins 122.

The thermal cycle recirculating pump 62 operates in two phases, the first of which is shown in

Figure 3. The cylindrical sleeve 110 is pressed against the cylindrical square valve seat 116 forming a pumping chamber 118 in the left annular chamber 113. The left exhaust valve 115 is open and the left outer section 112 has hot air applied by the heating means 123. The uncharged isotopes inside the annular chamber 113 are desorbed or vaporized and returned through the return pipe 114 to the discharge region 59.

There is an opening in the right annular chamber 113 which permits the randomly moving uncharged isotopes to enter that annular chamber 113. The exhaust valve 115 for the right annular chamber 113 is closed. The outer section 112 of the right annular chamber 113 is maintained at 200°C by cooling means 124. The uncharged isotopes are therefore devaporized as they strike the cooled outer section 112 of the right annular chamber 113.

As shown in Figure 4, after the first phase, the following changes occur for the second phase. The first cylindrical magnet 119 is deenergised and the second cylindrical magnet 120 is energized forcing the cylindrical sleeve 110 to the right. The left exhaust valve 115 is closed and the right exhaust valve 115 is opened. The cooling means 124 is connected to the left heat transfer fins 122 and the heating means 123 is connected to the right heat transfer fins 122.

The thermal cycle recirculating pump 62 continues operating, with the left annular chamber 113 devaporizing the randomly moving uncharged isotopes from the vacuum chamber 52. The right annular chamber 113 empties its contents by desorbing or vaporizing the condensed uncharged isotopes and returning them to the discharge region 59 through the return pipe 114.

Figures 3 and 4 disclose an embodiment of this invention useful with a condensable carrier gas such as bromine, chlorine, or fluorine. The uncharged isotope of the carrier gas will enter the thermal cycle recirculating pump 62 and be adsorbed and desorbed along with the uncharged isotope of iodine or other selected isotope.

#### Carrier Gas System

A third embodiment of the invention is disclosed in Figure 5. This shows a thermal cycle recirculating pump 62 suitable for a non-condensable carrier gas such as xenon. The thermal cycle recirculating pump 62 of Figure 5 is similar to the thermal cycle recirculating pump 62 of Figures 3 and 4, except that each annular chamber 113 has a second carrier gas exhaust valve 151 in addition to the exhaust valve 115 for iodine.

The temperature of the cooling means, e.g. 200°C, is chosen so that the walls of the annular

chambers 113 will not adsorb the non-condensable carrier gas xenon, but will adsorb the uncharged isotope of iodine. Therefore, the carrier gas alone will pass out of the open carrier gas exhaust valve.

When the cylindrical sleeve 110 seals an annular chamber 113, as it has the right annular chamber 113 in Figure 5, then the exhaust valve 115 for iodine is opened and the carrier gas exhaust valve is closed. The uncharged isotopes of iodine are desorbed by heating means 123 thereby recycling these isotopes.

#### Internal Baffle

A fourth embodiment of the invention is shown in Figure 6. This is a modification of the isotope purifier shown in Figure 5, with an internal baffle to reduce the escape of uncharged isotopes.

The internal baffle is shown as a cylindrical tube 130, attached to the second wall 88, which is not shown in Figure 5. The cylindrical tube 130 is mounted concentrically around aperture 99 in the second wall 88.

The cylindrical tube 130 allows the collimated ion beam to pass freely out of the vacuum chamber 52 and into the magnetic mass analyzer 54. The cylindrical tube 130 acts as an internal baffle because it traps the uncharged isotopes, returning them to the vacuum chamber 52 where they are recycled.

The cylindrical tube 130 traps the uncharged isotopes because the uncharged isotopes which randomly enter the cylindrical tube are then randomly reflected. The attenuation factor is approximately the ratio of the length to the internal diameter of the cylindrical tube 130.

The cylindrical tube 130 should have a diameter just large enough to pass the collimated ion beam. The length to diameter ratio is not critical, and may be a ratio of ten. The cylindrical tube 130 is heated to 600°C to prevent iodine from devaporizing. It may be made of high purity fused quartz.

The internal baffle may be of a different shape than cylindrical tube 130. If it is conical, approximating the divergence of the collimated ion beam, the attenuation will be even higher.

#### Other Embodiments

A turbine capable of operating at 600°C could serve as the recirculating pump for the uncharged isotopes. If the turbine wheels are made of glass or quartz, the iodine will not stick. There are at present turbo vacuum pumps designed to operate at these pressures. They have aluminum rotors, which would have to be modified.

Alternatively, a diffusion pump could serve as the recirculating pump. Diffusion pumps with glass, instead of metal, walls are known. The use of an oil diffusion pump would require a means to separate reactive isotopes such as iodine from the oil.

The walls of the vacuum chamber, and other parts, may be of Teflon coated metal rather than quartz. The Teflon coating would prevent the uncharged isotopes from adhering. This system would not require operation at elevated temperatures.

The thermal cycle recirculating pump of this invention could be used to implant ions onto a substrate. For instance rare ions could be implanted onto silicon to form an improved semiconductor. This invention would permit recirculation of the neutral particles to improve the yield and decrease the cost of the finished semiconductor.

It will thus be seen that the objects set forth above, among those made apparent from the preceding description, are efficiently attained and, since certain changes may be made in carrying out the above method and in the particular construction of the machine set forth without departing from the spirit and scope of the invention, it is intended that all matter contained in the above description and shown in the accompanying drawings shall be interpreted as illustrative and not in a limiting sense.

## Claims

1. A system for the separation and purification of a selected isotope from a polyisotopic mixture which comprises in combination:
  - a) an ion source (50) having a discharge region (59) containing means (60) to bombard the polyisotopic mixture to produce a mixture of ionized and uncharged isotopes;
  - b) a first aperture (58) from said discharge region (59) positioned to allow the ionized and uncharged isotopes to enter a vacuum chamber (52);
  - c) said vacuum chamber (52) having a plurality of walls (95; 112) maintained at an elevated temperature such that said uncharged isotopes are prevented from de-vaporizing;
  - d) a thermal cycle recirculating pump (62) connecting said vacuum chamber (52) and said discharge region (59) and having a selectively operable intake valve (142; 110) and a selectively operable exhaust valve (143; 115) and selectively operable heating means (150; 123) and selectively operable cooling means (150; 124);
  - e) said intake valve (142; 110) being open and said exhaust valve (143; 115) being closed during a first intake phase and being

reversed during a second exhaust phase;

f) said cooling means (150; 124) operable during said first phase and said heating means (150; 123) operable during said second phase;

g) said thermal cycle recirculating pump (62) having said uncharged isotopes de-vaporize and attach on the interior wall (95; 112) of the thermal cycle recirculating pump (62) during the first phase;

h) said thermal cycle recirculating pump (62) having said uncharged isotopes re-vaporize and return to the discharge region (59) during the second phase;

i) an extraction electrode means (92) adjacent said first aperture (58) to accelerate and direct said ionized isotopes toward a second aperture (99) as a collimated ion beam (61);

j) an analyser (54) to receive the collimated ion beam (61) of ionized isotopes and having a means (63) to separate the selected ionized isotope from the other ionized isotopes in the polyisotopic mixture; and

k) a plurality of collectors (64) to receive the isotopes, one of said collectors positioned to receive said selected isotope and the other collectors positioned to receive the other isotopes.

2. The isotope separation and purification system of claim 1 wherein:
  - a) a condensable carrier gas is introduced into the discharge region (59); and
  - b) means (145; 110, 119, 120) for allowing the carrier gas to pass into the thermal cycle recirculating pump (62) when the exhaust valve (143; 115) is closed and the intake valve (142) is open.
3. The isotope separation and purification system of claim 1 wherein:
  - a) the thermal cycle recirculating pump (62) includes a pair of annular chambers (113) disposed along the vacuum chamber (52);
  - b) each annular chamber (113) being disposed radially of said vacuum chamber (52);
  - c) each annular chamber (113) having its intake valve formed by a common sliding sleeve (110) and a cooperating pair of valve seats (116, 117); and
  - d) each annular chamber (113) having at least one selectively operable exhaust valve (115), a selectively operable heating means (123), and a selectively operable cooling means (124).

4. The isotope separation and purification system of claim 3 wherein:
- a) a noncondensable carrier gas is introduced into the discharge region (59); and
  - b) a second exhaust valve (151) for each annular chamber (113) is positioned to allow the carrier gas to pass through the annular chamber (113) when a first exhaust valve (115) is closed and the sleeve valve (110) is separated from its associated valve seat (116, 117).
5. The isotope separation and purification system of claim 1 wherein:
- a) said vacuum chamber (52) contains an internal baffle (130);
  - b) said internal baffle (130) being generally cylindrical in shape;
  - c) one end of the internal baffle (130) being attached to the second aperture (99) of the vacuum chamber (52);
  - d) said internal baffle (130) being aligned to surround the collimated beam (61) of ionized particles;
  - e) said internal baffle (130) attenuating the random discharge of uncharged isotopes through the second aperture (99).
6. In a method for the separation and purification of a selected isotope from a polyisotopic mixture having the following steps:
- a) bombarding the polyisotopic mixture in an ion source (50) to produce ionized particles;
  - b) accelerating said ionized particles out of the ion source (50) and directing them to an ion deposit location;
  - c) allowing the neutral particles to randomly leave the ion source (50);
  - d) passing the ionized particles through, and the neutral particles into, a vacuum chamber (52), as they leave the ion source (50);
  - e) maintaining the vacuum chamber (52) at a temperature above the vaporization temperature of the neutral particles;
  - f) allowing the neutral particles to exit the vacuum chamber (52) into a thermal cycle recirculating pump (62);
  - g) cooling and devaporizing the neutral particles in the thermal cycle recirculating pump (62);
  - h) heating and revaporizing the devaporized neutral particles in the thermal cycle recirculating pump (62); and
  - i) recycling the revaporized neutral particles to the ion source (50) where they are again bombarded to produce additional ionized particles.

## Patentansprüche

1. System zum Trennen und Reinigen eines aus einem Polyisotopengemisch gewählten Isotops, welches folgendes in Kombination aufweist:
- a) eine Ionenquelle (50) mit einem Entladungsbereich (59), der eine Einrichtung (60) aufweist, um das Polyisotopengemisch zur Gewinnung eines Gemischs aus ionisierten und ungeladenen Isotopen zu beschleunigen;
  - b) eine erste aus dem Entladungsbereich (59) führende Öffnung (58), die so positioniert ist, daß die ionisierten und ungeladenen Isotopen in eine Unterdruckkammer (52) übertreten können,
  - c) wobei die Unterdruckkammer (52) eine Vielzahl von Wandungen (95; 112) aufweist, die auf erhöhter Temperatur so gehalten werden, daß eine Entnebelung der ungeladenen Isotopen verhindert wird;
  - d) eine Umwälzpumpe (62) mit thermischer Wechselbelastung, welche die Unterdruckkammer (52) mit dem Entladungsbereich (59) verbindet und ein selektiv betätigbares Einlaßventil (142; 110) und ein selektiv betätigbares Auslaßventil (143; 115) sowie selektiv betätigbare Heizeinrichtungen (150; 123) und selektiv betätigbare Kühleinrichtungen (150; 124) aufweist,
  - e) wobei während einer ersten Einlaßphase das Einlaßventil (142; 110) geöffnet und das Auslaßventil (143; 115) geschlossen ist und beide während einer zweiten Auslaßphase umgekehrt geschaltet sind;
  - f) wobei die Kühleinrichtung (150; 124) während der ersten Phase und die Heizeinrichtung (150; 123) während der zweiten Phase betreibbar sind;
  - g) wobei die Umwälzpumpe (62) mit thermischer Wechselbelastung während der ersten Phase die ungeladenen Isotopen entnebeln und sich an ihren Innenwandungen (95; 112) festsetzen läßt;
  - h) wobei die Umwälzpumpe (62) mit thermischer Wechselbelastung während der zweiten Phase die ungeladenen Isotopen erneut verdampfen und in den Entladungsbereich (59) zurückkehren läßt;
  - i) eine Elektrodeneinrichtung (92) zur Extraktion, die sich nahe der ersten Öffnung (58) befindet, um die ionisierten Isotopen zu beschleunigen und in Form eines kollimierten Ionenstrahls (61) zu einer zweiten Öffnung zu lenken;
  - j) einen Analysator (54) zum Empfangen des kollimierten Ionenstrahls (61) aus ionisierten Isotopen, welcher eine Einrichtung



- (63) zum Abtrennen der ausgewählten ionisierten Isotopen von den anderen ionisierten Isotopen in dem Polyisotopengemisch aufweist, und
- k) eine Vielzahl von Kollektoren (64) zum Auffangen der Isotopen, wobei einer der Kollektoren so positioniert ist, daß er das ausgewählte Isotop empfängt, während die anderen Kollektoren so positioniert sind, daß sie die anderen Isotopen empfangen.
2. System zum Trennen und Reinigen von Isotopen nach Anspruch 1, bei welchem:
- a) ein kondensierbares Trägergas in den Entladungsbereich (59) eingeleitet wird, und
- b) eine Einrichtung (145; 110, 119, 120) vorgesehen ist, welche das Trägergas in die Umwälzpumpe mit thermischer Wechselbelastung (62) übertreten läßt, wenn das Auslaßventil (143; 115) geschlossen und das Einlaßventil (142) offen ist.
3. System zum Trennen und Reinigen von Isotopen nach Anspruch 1, bei welchem:
- a) die Umwälzpumpe mit thermischer Wechselbelastung (62) ein Paar Ringkammern (113) aufweist, welche entlang der Unterdruckkammer (52) angeordnet sind,
- b) wobei jede Ringkammer (113) bezüglich der Unterdruckkammer (52) radial angeordnet ist,
- c) an jeder Ringkammer (113) eine gemeinsame Schiebehülse (110) und ein damit zusammenwirkendes Paar Ventilsitze (116, 117) deren Einlaßventil bildet, und
- d) bei welchem jede Ringkammer (113) mindestens ein selektiv betätigbares Auslaßventil (115), eine selektiv betätigbare Heizeinrichtung (123) und eine selektiv betätigbare Kühleinrichtung (124) aufweist.
4. System zum Trennen und Reinigen von Isotopen nach Anspruch 3, bei welchem:
- a) ein nicht kondensierbares Trägergas in den Entladungsbereich (59) eingeleitet wird, und
- b) ein zweites Auslaßventil (151) für jede Ringkammer (113) so positioniert ist, daß es das Trägergas durch die Ringkammer (113) in die Umwälzpumpe mit thermischer Wechselbelastung (62) übertreten läßt, wenn ein erstes Auslaßventil (115) geschlossen und das hülsenförmige Ventil (110) vom zugeordneten Ventilsitz (116, 117) getrennt ist.
5. System zum Trennen und Reinigen von Isotopen nach Anspruch 1, bei welchem:
- a) die Unterdruckkammer (52) eine innenliegende Leiteinrichtung (130) enthält;
- b) die innenliegende Leiteinrichtung (130) im wesentlichen zylinderförmig ist;
- c) ein Ende der innenliegenden Leiteinrichtung (130) an der zweiten Öffnung (99) der Unterdruckkammer (52) angebracht ist;
- d) die innenliegende Leiteinrichtung (130) so ausgerichtet ist, daß sie den kollimierten Strahl (61) aus ionisierten Teilchen umschließt,
- e) wobei die innenliegende Leiteinrichtung (130) die zufallsbedingte Entladung ungeladener Isotopen durch die zweite Öffnung (99) dämpft.
6. Bei einem Verfahren zum Trennen und Reinigen eines aus einem Polyisotopengemisch gewählten Isotops, welches folgende Schritte aufweist:
- a) Beschießen des Polyisotopengemisches in einer Ionenquelle (50) zur Gewinnung ionisierter Teilchen;
- b) Beschleunigen der ionisierten Teilchen aus der Ionenquelle (50) heraus und Lenken derselben zu einer Ionenspeicherstelle;
- c) Ermöglichen des Austritts neutraler Teilchen in zufälliger Weise aus der Ionenquelle (50);
- d) Überleiten der ionisierten Teilchen durch eine Unterdruckkammer (52) hindurch und Einleiten der neutralen Teilchen in die Unterdruckkammer (52) bei deren Austritt aus der Ionenquelle (50);
- e) Halten der Unterdruckkammer (52) auf einer Temperatur oberhalb der Verdampfungstemperatur der neutralen Teilchen;
- f) Gestatten des Austritts der neutralen Teilchen aus der Unterdruckkammer (52) in eine Umwälzpumpe mit thermischer Wechselbelastung (62);
- g) Abkühlen und Entnebeln der neutralen Teilchen in der Umwälzpumpe mit thermischer Wechselbelastung (62);
- h) Erwärmen und erneutes Verdampfen der entnebelten neutralen Teilchen in der Umwälzpumpe mit thermischer Wechselbelastung (62), und
- i) Rückführen der erneut verdampften neutralen Teilchen in die Ionenquelle (50), wo sie erneut zur Gewinnung weiterer ionisierter Teilchen beschossen werden.

## Revendications

1. Système pour la séparation et la purification d'un isotope choisi, à partir d'un mélange polyisotopique, qui comprend en combinaison ;

a) une source d'ions (50) possédant une région de décharge (59) contenant des moyens (60) pour bombarder le mélange polyisotopique afin de produire un mélange d'isotopes ionisés et non chargés ; 5

b) une première ouverture (58) de la région de décharge (59), positionnée pour permettre aux isotopes ionisés et non chargés de pénétrer dans une chambre à vide (52) ; 10

c) la chambre à vide (52) comportant une multiplicité de parois (95 ; 112) maintenues à une température élevée de telle sorte que les isotopes non chargés sont empêchés de se dévaporer ; 15

d) une pompe de recirculation à cycle thermique (62) mettant en communication la chambre à vide (52) et la région de décharge (59) et comportant une soupape d'admission actionnable sélectivement (142 ; 110), une soupape d'échappement actionnable sélectivement (153 ; 115), des moyens de chauffage actionnables sélectivement (150 ; 123) et des moyens de refroidissement actionnables sélectivement (150 ; 124) ; 20

e) la soupape d'admission (142 ; 110) étant ouverte et la soupape d'échappement (143 ; 115) étant fermée pendant une première phase d'admission et étant inversée pendant une seconde phase d'échappement ; 25

f) les moyens de refroidissement (150 ; 124) étant actionnables pendant la première phase et les moyens de chauffage (150 ; 123) étant actionnables pendant la seconde phase ; 30

g) la pompe de recirculation à cycle thermique (62) faisant en sorte que les isotopes non chargés se dévaporent et s'attachent à la paroi intérieure (95 ; 112) de la pompe de recirculation à cycle thermique (62) pendant la première phase ; 35

h) la pompe de recirculation à cycle thermique (62) faisant en sorte que les isotopes non chargés se revaporent et retournent à la région de décharge (59) pendant la seconde phase ; 40

i) Moyen d'électrode d'extraction (92) adjacent à la première ouverture (58) pour accélérer et diriger les isotopes ionisés vers une seconde ouverture (99) sous forme d'un faisceau collimaté d'ions (61) ; 45

j) un analyseur (54) pour recevoir le faisceau collimaté d'ions (61) d'isotopes ionisés et comportant un moyen (63) pour séparer l'isotope ionisé choisi des autres isotopes ionisés du mélange polyisotopique ; et 50

k) une multiplicité de collecteurs (64) propres à recevoir les isotopes, l'un des collec-

teurs étant positionné pour recevoir l'isotope choisi et les autres collecteurs étant positionnés pour recevoir les autres isotopes.

2. Système de séparation et de purification d'isotope selon la revendication 1 dans lequel :
  - a) un gaz porteur condensable est introduit dans la région de décharge (59) ; et
  - b) des moyens (145 ; 110, 119, 120) pour permettre au gaz porteur de passer dans la pompe de recirculation à cycle thermique (62) lorsque la soupape d'échappement (143 ; 115) est fermée et la soupape d'admission (142) est ouverte.
3. Système de séparation et de purification d'isotope selon la revendication 1 dans lequel :
  - a) la pompe de recirculation à cycle thermique (62) comprend une paire de chambres annulaires (113) disposées le long de la chambre à vide (52) ;
  - b) chaque chambre annulaire (113) étant disposée radialement par rapport à la chambre à vide (52) ;
  - c) chaque chambre annulaire (113) ayant sa soupape d'admission formée par un manchon coulissant commun (110) et une paire coopérante de sièges de soupape (116, 117) ; et
  - d) chaque chambre annulaire (113) comportant au moins une soupape d'échappement (115) actionnable sélectivement, des moyens de chauffage (123) actionnables sélectivement, et des moyens de refroidissement (124) actionnables sélectivement.
4. Système de séparation et de purification d'isotope selon la revendication 3, dans lequel :
  - a) un gaz porteur non condensable est introduit dans la région de décharge (59) ; et
  - b) une seconde soupape d'échappement (151) pour chaque chambre annulaire (113) est positionnée de manière à permettre au gaz porteur de passer au travers de la chambre annulaire (113) lorsqu'une première soupape d'échappement (115) est fermée et la soupape en forme de manchon (110) est séparée de son siège de soupape associé (116, 117).
5. Système de séparation et de purification d'isotope selon la revendication 1, dans lequel :
  - a) la chambre à vide (52) contient une cloison interne (130) ;
  - b) la cloison interne (130) étant de forme généralement cylindrique ;
  - c) une extrémité de la cloison interne (130) étant attachée à la seconde ouverture (99)

de la chambre sous vide (52) ;

d) la cloison interne (130) étant alignée pour entourer le faisceau collimaté (61) de particules ionisées ;

e) la cloison interne (130) atténuant la décharge au hasard des isotopes non chargés au travers de la seconde ouverture (99). 5

6. Procédé pour la séparation et la purification d'un isotope choisi à partir d'un mélange polyisotopique, comprenant les opérations suivantes : 10

a) bombarder le mélange polyisotopique dans une source d'ions (50) pour produire des particules ionisées ; 15

b) accélérer les particules ionisées hors de la source d'ions (50) et les diriger vers un endroit de dépôt d'ions ;

c) permettre aux particules neutres de quitter au hasard la source d'ions (50) ; 20

d) passer les particules ionisées à travers une chambre à vide (52) et les particules neutres dans cette chambre à vide, à mesure qu'elles quittent la source d'ions (50) ;

e) maintenir la chambre à vide (52) à une température supérieure à la température de vaporisation des particules neutres ; 25

f) permettre aux particules neutres de sortir de la chambre à vide (52) et de pénétrer dans une pompe de recirculation à cycle thermique (62) ; 30

g) refroidir et évaporer les particules neutres dans la pompe de recirculation à cycle thermique (62) ;

h) chauffer et revaporer les particules neutres évaporisées dans la pompe de recirculation à cycle thermique (62) ; et 35

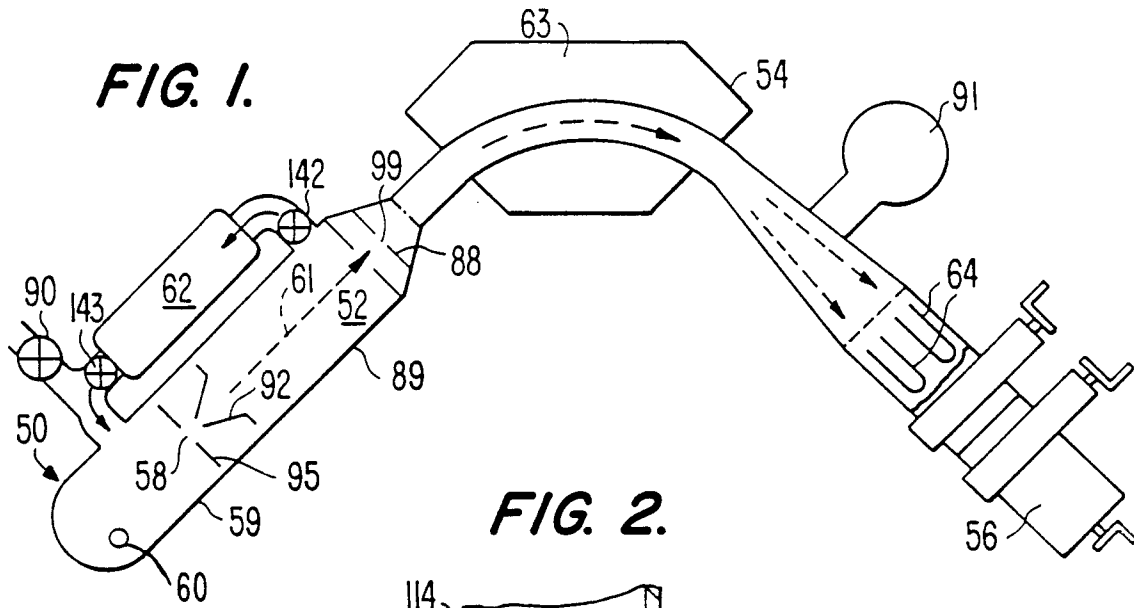
i) recycler les particules neutres revaporisées vers la source d'ions (50) où elles sont à nouveau bombardées pour produire des particules ionisées additionnelles. 40

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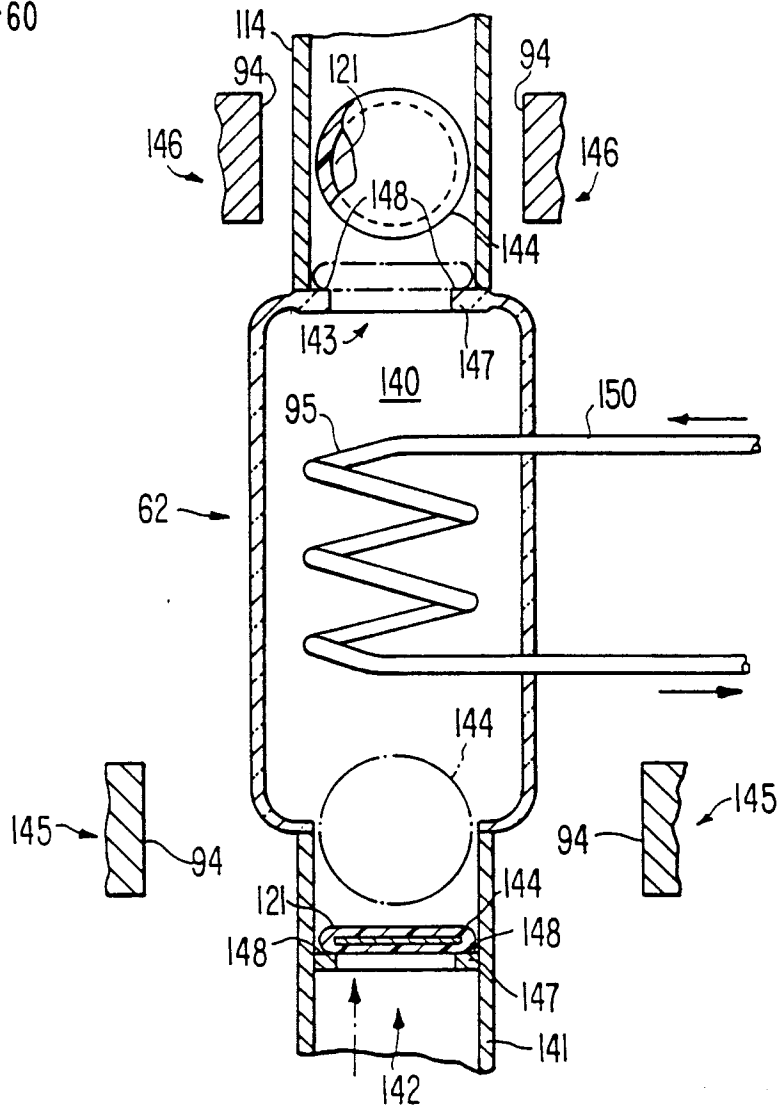
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**FIG. 1.**



**FIG. 2.**



**FIG. 3.**

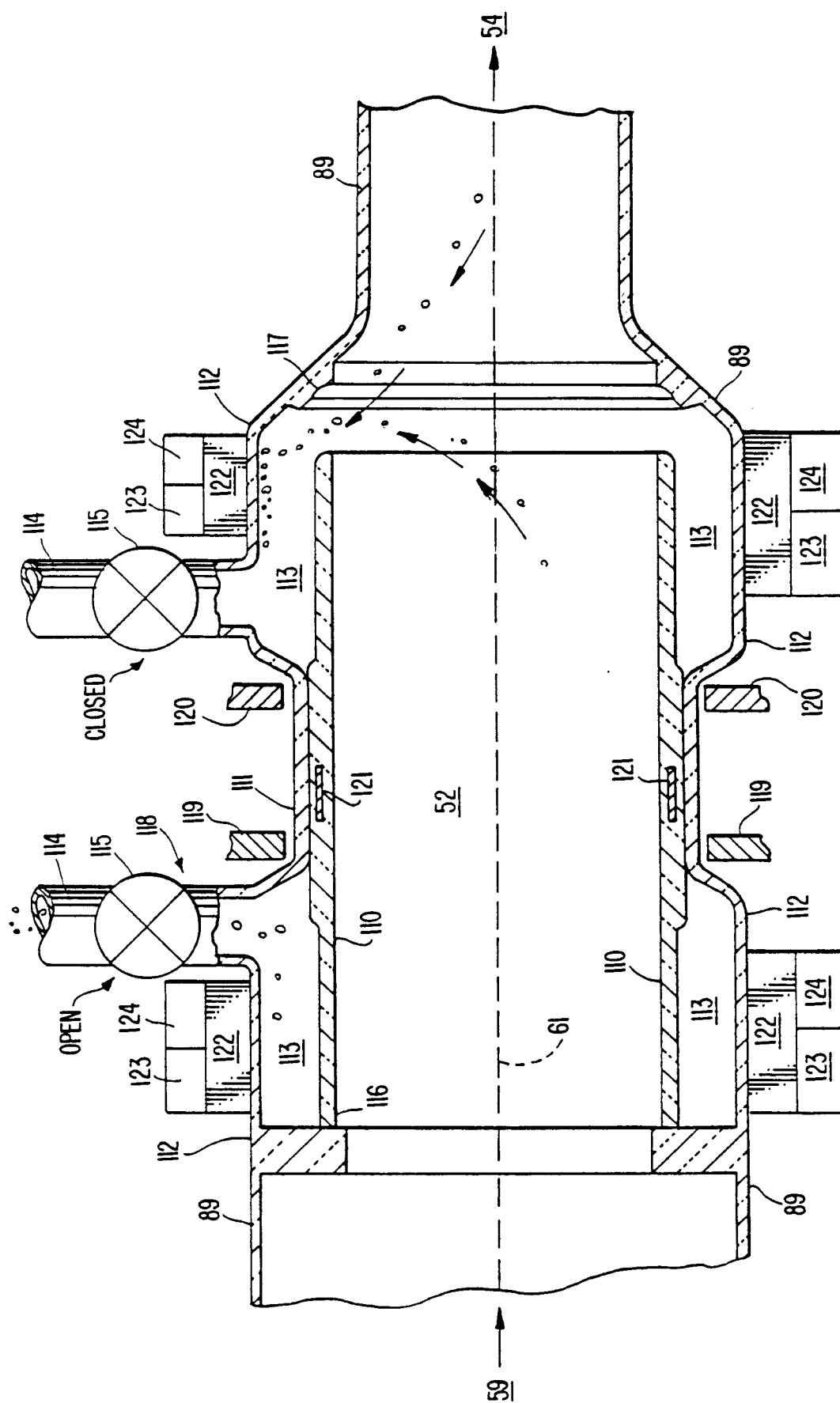
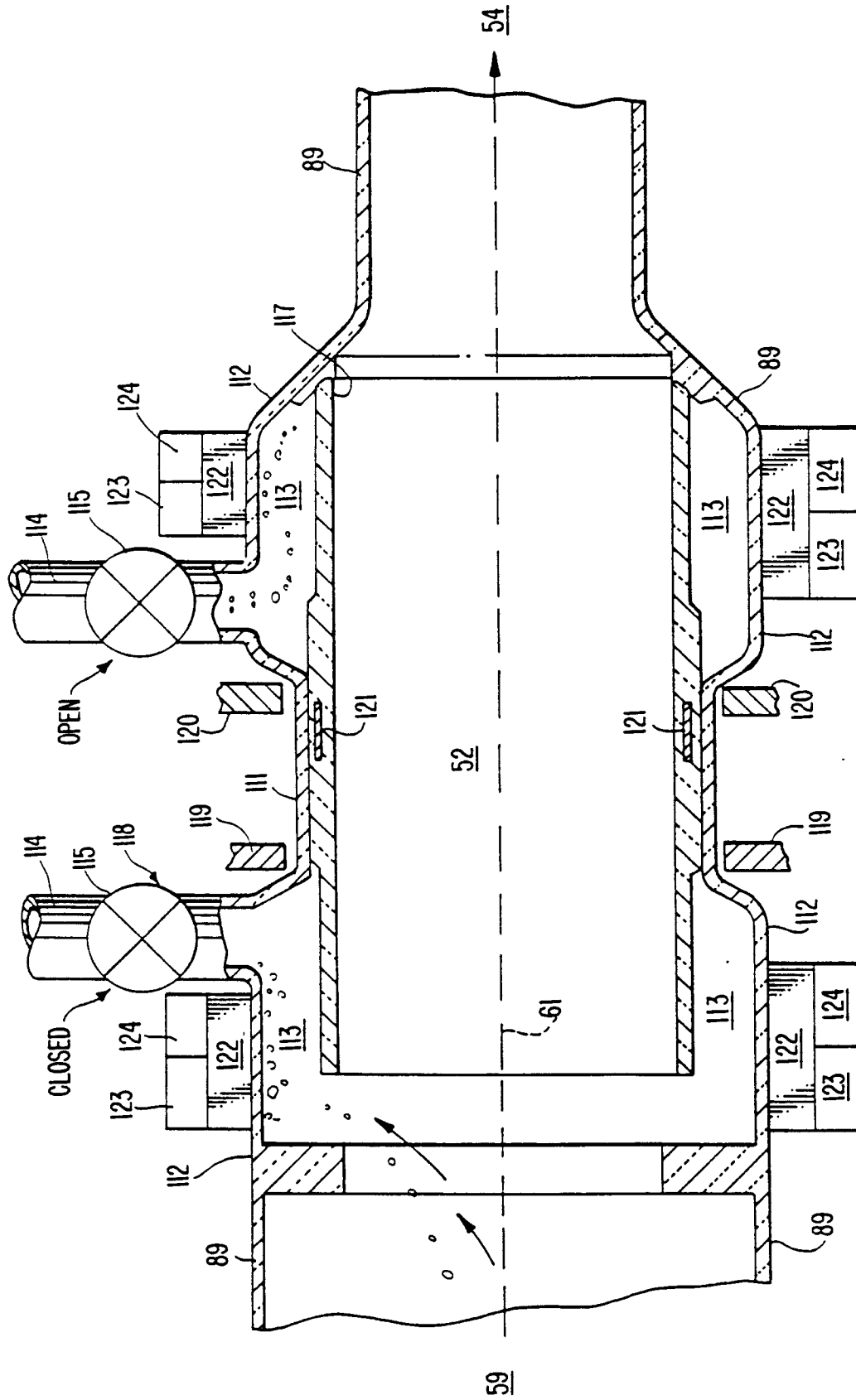


FIG. 4.



**FIG. 5.**

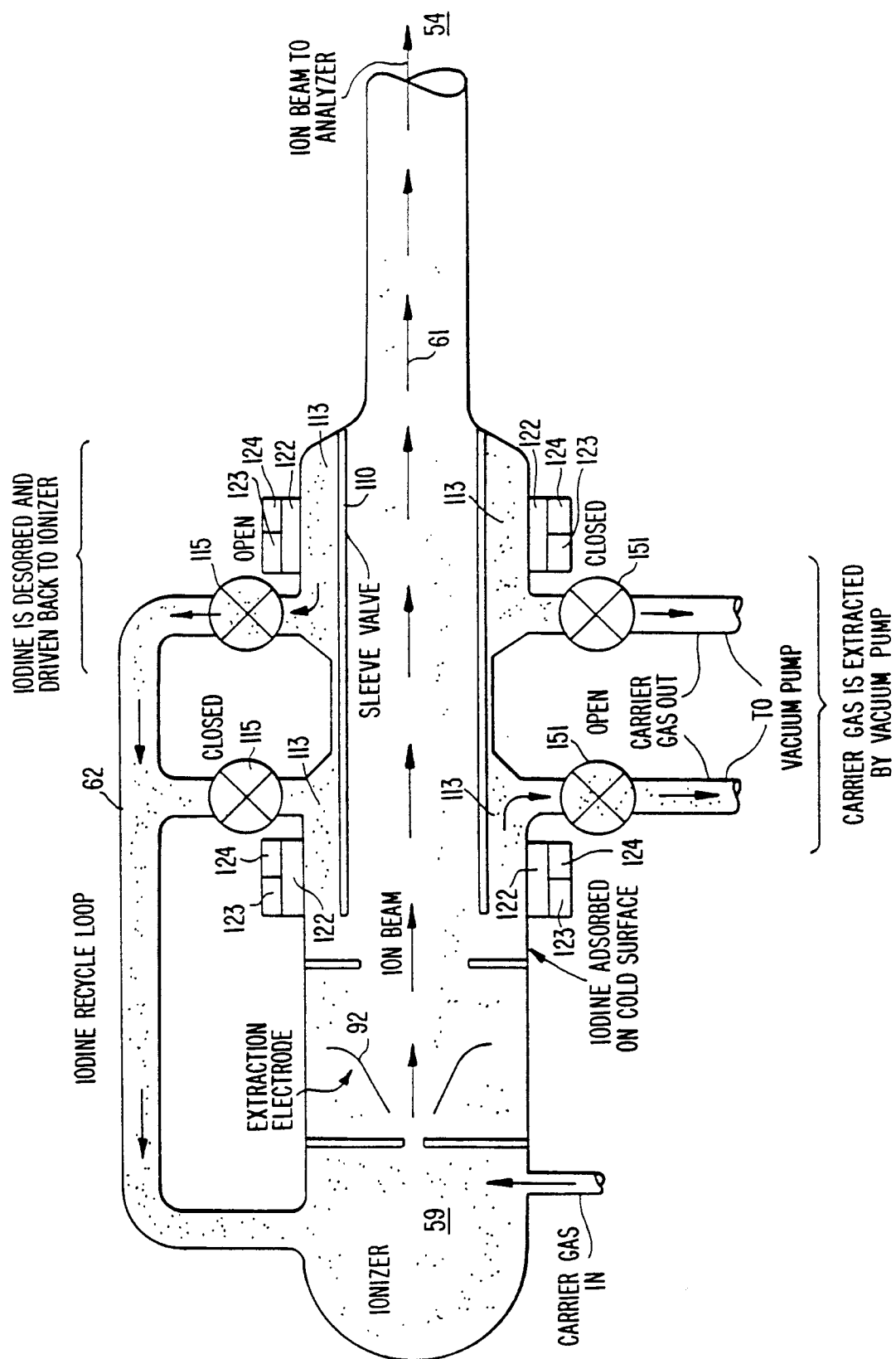


FIG. 6.

